

Single Photon Generation from Nitrogen Atomic-Layer Doped Gallium Arsenide

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Abstract. We have studied the properties of photoluminescence (PL) from individual isoelectronic traps formed by nitrogen-nitrogen (NN) pairs in nitrogen atomic-layer doped (ALD) GaAs. Micro-PL measurements were performed to investigate the properties of single photons generated from individual isoelectronic traps. Twin PL peaks were observed from individual isoelectronic traps in nitrogen ALD GaAs(001). The PL transitions at longer and shorter wavelength sides were linearly polarized in the [110] and [1-10] directions, respectively. The peak splitting and polarization properties can be explained by some in-plane anisotropy most likely due to strain in host crystal. From individual isoelectronic traps in nitrogen ALD GaAs(111), a single PL peak with random polarization was observed, showing that the growth on (111) surface is an effective way to obtain unpolarized single photons. As for nitrogen ALD GaAs(110), different polarization properties were obtained depending on the atomic configuration of NN pairs. In addition, we have used AlGaAs layers to diminish the in-plane anisotropy and could successfully obtain single emission lines with unpolarized character. Introducing AlGaAs layers was also useful for improving the luminescence efficiency.

Introduction

Quantum key distribution (QKD) guarantees secure communication of a private key based on the foundations of quantum mechanics. BB84 protocol [1] is a QKD scheme that uses the polarization of single photons. Thus, single photon generation is expected to play an essential role in QKD. In order to realize single photon generation, several candidates are proposed, for example, acceptors in semiconductors [2], nitrogen-vacancy states in diamond [3], and so on. Above all, semiconductor quantum dots [4] are intensively studied for single photon generation. In addition to these ways, the use of isoelectronic traps [5-9] is also a promising candidate for single photon generation. In nitrogen doped GaAs, since the electronegativity of nitrogen is larger than that of arsenic, individual nitrogen-nitrogen (NN) pairs act as isoelectronic traps to which only one electron, or only one exciton is bound. The wavelengths of the emission due to isoelectronic traps is determined by the atomic configuration of NN pairs [10]. For example, the emission observed at 868 nm is assigned to the nearest neighbor NN pair labeled NN_B. In addition, the emission linewidth is extremely narrow. Thus, sharp emission lines with highly-reproducible wavelengths are easily obtained from isoelectronic

traps due to NN pairs in GaAs. This is advantageous to the design of distributed Bragg reflectors for enhancing or selecting photons with specific wavelengths. In the present paper, we report polarization properties of PL from individual isoelectronic traps formed by NN pairs in metalorganic vapor phase epitaxy (MOVPE)-grown nitrogen atomic-layer doped (ALD) GaAs and GaAs/AlGaAs heterostructures.

Experimental Procedures

The samples used in this study were nitrogen ALD GaAs and GaAs/AlGaAs heterostructures grown by low-pressure MOVPE. The substrates used were semi-insulating undoped GaAs(001), (111)A, and (110). Trimethylgallium (TMG), trimethylaluminum (TMA), tertiarybutylarsine (TBA), and dimethylhydrazine (DMHy) were used as the Ga, Al, As, and N precursors, respectively. The growth temperatures were between 600 and 650°C. As an example, Fig. 1 shows the time sequence for the growth of nitrogen ALD GaAs/AlGaAs. To perform nitrogen atomic-layer doping into GaAs, we supplied DMHy flow during the interruption of TMG flow for several seconds. The nitrogen ALD GaAs/AlGaAs doubleheterostructures consists of a 40-nm thick nitrogen ALD GaAs layer and 50-nm thick AlGaAs barrier layers. Nitrogen atomic-layer doping was carried out at the center of the GaAs layer.

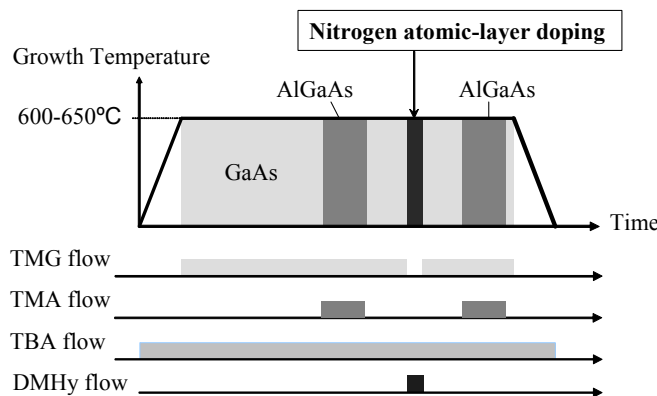


Fig. 1 Time sequence for the growth of nitrogen ALD GaAs/AlGaAs heterostructures. Nitrogen atomic-layer doping is carried out by supplying DMHy flow during the interruption of TMG flow.

We have measured micro-PL spectra at 4 K using a diode-pumped solid-state laser (532 nm) as the excitation source to study the properties of single photons generated from individual isoelectronic traps. The luminescence was dispersed by a 0.75-m monochromator and detected by an intensified charge-coupled device. The spatial resolution and energy resolution of the micro-PL measurement system were $\sim 1 \mu\text{m}$ and $\sim 30 \mu\text{eV}$, respectively. We have also carried out polarization measurements of PL spectra.

Results and Discussion

Nitrogen ALD GaAs on (001) A micro-PL intensity map of nitrogen ALD GaAs grown on a GaAs(001) substrate is shown in Fig. 2. The map was made by scanning the sample in one direction. As can be seen from this figure, several twin emission lines with narrow linewidths are observed at specific positions. This indicates that the emission from individual isoelectronic traps could be successfully obtained. Twin emission lines located at 868 nm, 856 nm, and 850 nm are assigned to NN pair labeled NN_B , NN_D , and NN_E [10], respectively. So far, twin emission observed at 852 nm has not been identified. However, several characters, i.e., localized, narrow and twin, suggest that it is also due to an isoelectronic trap formed by a NN pair.

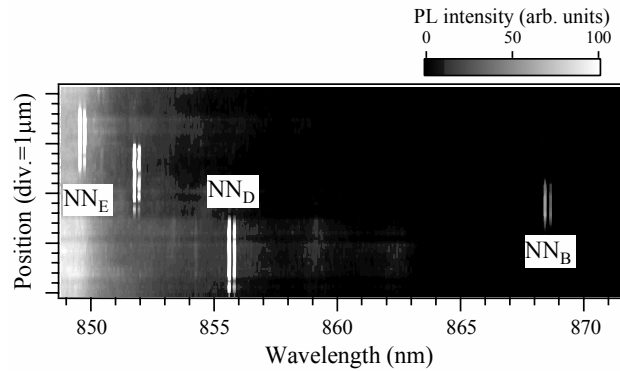


Fig. 2 Micro-PL intensity map of nitrogen ALD GaAs grown on a (001) substrate.

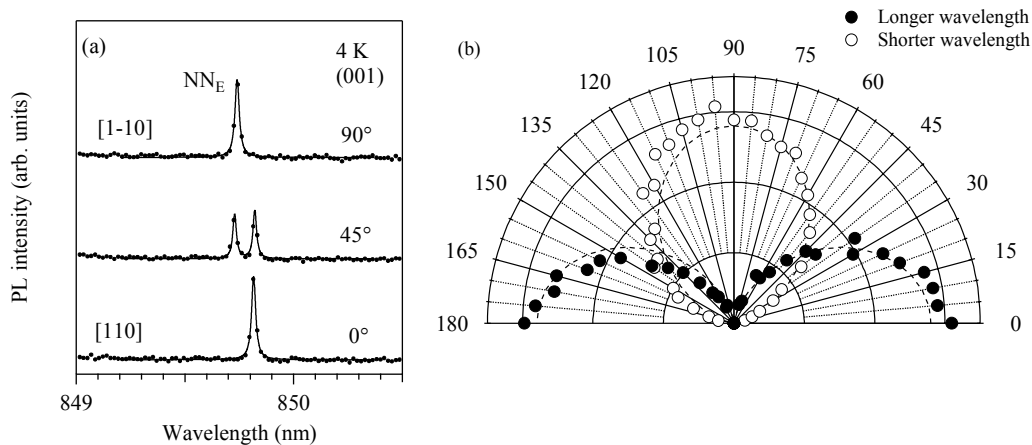


Fig. 3 (a) Polarized PL spectra from an individual isoelectronic trap formed by a NN pair labeled NN_E in nitrogen ALD GaAs grown on a (001) substrate. (b) PL peak intensities as a function of polarization angle.

Fig. 3 (a) shows polarized PL spectra obtained from an individual isoelectronic trap formed by a NN pair labeled NN_E in nitrogen ALD GaAs grown on a (001) substrate. For the [110] polarization, only the emission line at the longer wavelength side is seen, while only shorter wavelength emission is observed for the [1-10] polarization. Fig. 3 (b) shows the PL intensities as a function of polarization angle, clearly indicating that the emission lines at the longer and shorter wavelength side are linearly polarized along the [110] and [1-10] directions, respectively. The same polarization property is seen for all the twin emission lines in nitrogen ALD GaAs grown on (001) substrates. This polarization property suggests that the level splitting is due to not the arrangement of NN pairs but anisotropy in the host crystal, probably strain anisotropy in the sample. Similar polarization properties have been reported for Te ALD ZnS [11]; the shorter and longer wavelength emission peaks are linearly polarized along the [110] and [1-10] directions, respectively. In this case, almost all twins follow the identical behavior, and the splitting is explained by the strain field created by Te atoms. Considering a similar situation, nitrogen atoms with a smaller ionic radius probably cause the strain anisotropy between the [110] and [1-10] directions in the sample.

Nitrogen ALD GaAs on (111) Fig. 4 (a) shows a PL spectrum from an individual isoelectronic trap formed by a NN pair labeled NN_D in nitrogen ALD GaAs grown on a (111) substrate. As can be seen from the PL spectrum, single peak emission is obtained unlike twin PL peaks observed from nitrogen ALD GaAs grown on (001) substrates. When (111) substrates are used, emission lines observed from individual isoelectronic traps due to NN pairs other than NN_D always show a single-peak character. Fig. 4 (b) shows the PL peak intensity as a function of polarization angle. The almost constant PL intensity at any polarization angle indicates random polarization or circular polarization. We have measured polarized PL intensity using a quarter-wave plate and a polarization analyzer, and confirmed that the PL is not circularly polarized but randomly polarized. Thus, it was found from the polarization measurements that randomly polarized single photons desirable for the application to QKD can be obtained. This result shows that in-plane anisotropy does not exist in the case of using (111) substrates, which is reasonable taking into consideration the crystal symmetry.

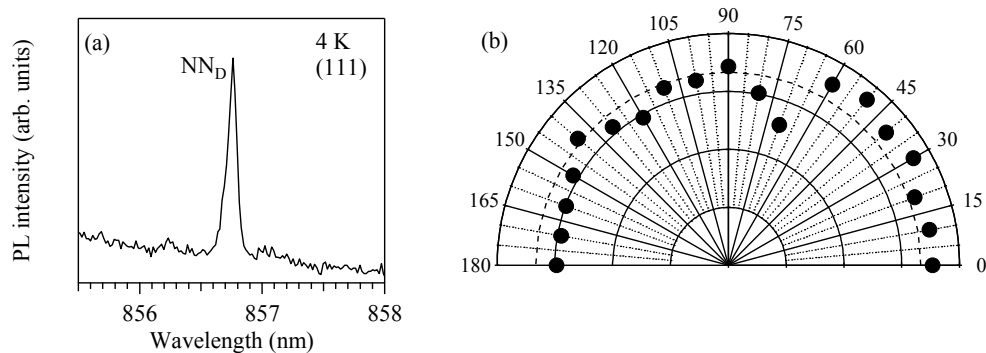


Fig. 4 (a) PL spectrum of an individual isoelectronic trap labeled NN_E in nitrogen ALD GaAs grown on a (111) substrate. (b) PL peak intensity as a function of polarization angle.

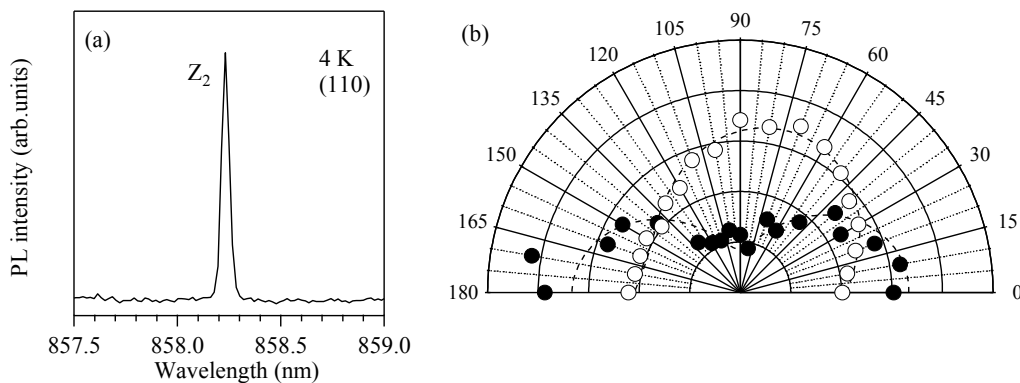


Fig. 5 (a) PL spectrum of an individual isoelectronic trap labeled Z_2 in nitrogen ALD GaAs grown on a (110) substrate. (b) Polarization angle dependence of the PL intensity of the same kind of isoelectronic traps located at different positions.

Nitrogen ALD GaAs on (110) Fig. 5 (a) shows a PL spectrum of an individual isoelectronic trap formed by a NN pair labeled Z_2 in nitrogen ALD GaAs grown on a (110) substrate. Although the isoelectronic trap labeled Z_2 shows a single peak emission, splitting PL peaks were sometimes observed for other kinds of NN pairs. Fig. 5 (b) shows the polarization angle dependence of the PL intensity of isoelectronic traps labeled Z_2 located at different positions. The luminescence of one isoelectronic trap Z_2 tends to be polarized along the [001] direction, while the luminescence of the other Z_2 is polarized along the [1-10] direction. In addition, different polarization properties were obtained depending on the atomic configuration of NN pairs. Similar results have been reported for nitrogen ALD GaP, and the polarization is explained by the pairing direction of isoelectronic traps

formed by two nitrogen atoms [8]. Thus, the polarization properties of individual isoelectronic traps in nitrogen ALD GaAs grown on (110) are probably determined by the atomic configuration of NN pairs, unlike nitrogen ALD GaAs grown on (001) or (111).

Nitrogen ALD GaAs/AlGaAs heterostructure As discussed above, in-plane strain anisotropy affects the splitting and polarization properties of individual isoelectronic traps in nitrogen ALD GaAs on (001). Thus, we tried to diminish the in-plane strain anisotropy by sandwiching a nitrogen ALD layer between AlGaAs which has a little larger lattice constant than GaAs. In Fig. 6 (a), a sharp single PL peak is observed from an individual isoelectronic trap due to a NN pair labeled Z_1 in the nitrogen ALD GaAs/AlGaAs heterostructure grown on GaAs(001). Fig. 6 (b) shows the PL peak intensity as a function of polarization angle. The intensity is found to be almost constant at any polarization angle, indicating that random polarization can be obtained. This result suggests that AlGaAs layers can diminish the in-plane strain anisotropy and result in the single peak and random polarization.

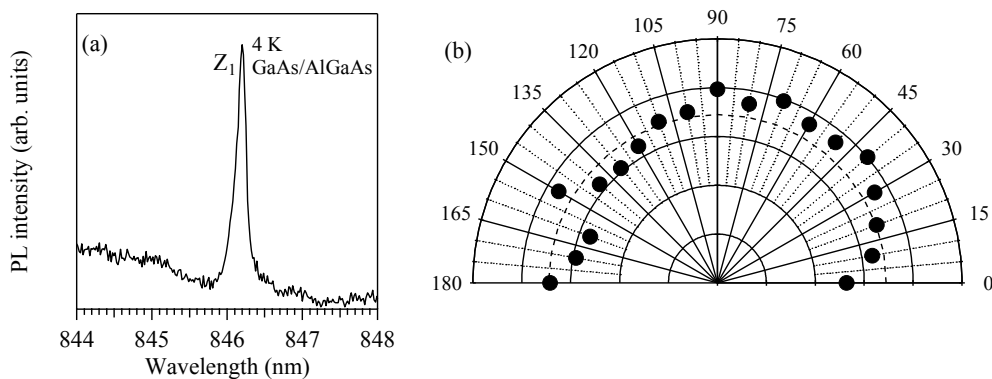


Fig. 6 (a) PL spectrum of an individual isoelectronic trap labeled Z_1 in a nitrogen ALD GaAs/GaAs(001) heterostructure. (b) PL peak intensity as a function of polarization angle.

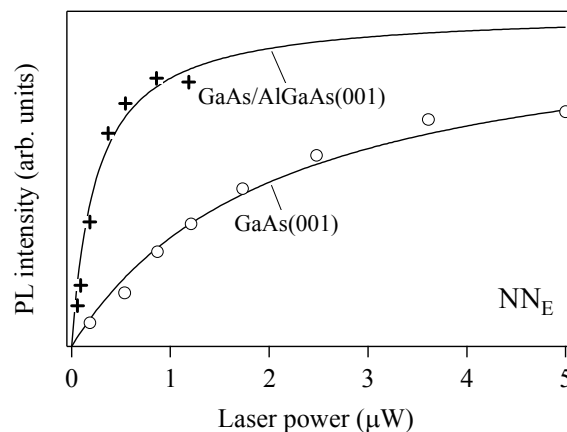


Fig. 7 Excitation power dependence of the PL intensity of an individual isoelectronic trap labeled NN_E in nitrogen ALD GaAs on a (001) substrate and a nitrogen ALD GaAs/AlGaAs(001) heterostructure.

Since we can expect that the introduction of AlGaAs layers improves the luminescence efficiency, the excitation power dependence of the emission was investigated to clarify the effect. Fig. 7 shows the laser power dependence of the PL intensity of an individual isoelectronic trap labeled NN_E in nitrogen ALD GaAs on a (001) substrate and a nitrogen ALD GaAs/AlGaAs(001) heterostructure measured under the same conditions. With increasing laser power, as can be seen from this figure, the

PL intensity rapidly increases at lower powers, and gradually becomes constant. Compared to nitrogen ALD GaAs, the increasing rate of the PL intensity for nitrogen ALD GaAs/AlGaAs is approximately 8 times larger at lower laser power. This is attributed to the confinement effect of AlGaAs barrier layers. Therefore, AlGaAs layers are useful for not only obtaining unpolarized single photons but also improving the luminescence efficiency.

Summary

We studied the PL from individual isoelectronic traps formed by NN pairs in nitrogen ALD GaAs grown on GaAs(001), (111) and (110) substrates. Twin PL peaks were observed from individual isoelectronic traps in nitrogen ALD GaAs on (001). Longer and shorter wavelength emission lines were linearly polarized in the [110] and [1-10] directions, respectively. From individual isoelectronic traps in nitrogen ALD GaAs on (111), a single PL peak showing unpolarized character was observed. The polarization properties of the emission from individual isoelectronic traps in nitrogen ALD GaAs on (110) were dependent of the atomic configuration of NN pairs. In addition, we tried to diminish the in-plane strain anisotropy by sandwiching a nitrogen ALD GaAs between AlGaAs and could successfully observed unpolarized single emission lines. The use of AlGaAs was also useful for improving the luminescence efficiency.

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